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Environmental effects on quantum relaxation and coherent dynamics in rare-earths ions

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Abstract

This work provides new developments of the field of spin dynamics in mesoscopic systems. We first discuss, by a few examples, how environmental degrees of freedom affect tunneling and subsequent slow quantum relaxation of large molecule spins (such as Mn_{12}–AC) or rare-earth (RE) spins (such as Ho diluted in $YLiF_4$) showing two different facets of the spin bath model. The case of low molecule spins (V_{15}) is also considered. Then we discuss AC-susceptibility experiments performed on $YLiF_4$:Ho^{3+} allowing to clarify the roles played by the phonon and the spin baths on single-ion and two-ion electro-nuclear tunneling. In the last part, we extend these quantum dynamical studies to the case of Er diluted in $CaWO_4$ where the spin and phonon baths are weak enough to allow fast and coherent quantum dynamics with the observation of Rabi oscillations. The angular momentum and magnetic moment of Er, $J = 15/2$and $9\,\mu_B$, are comparable to those of large molecule spins, implying easy manipulations of potential RE solid-state qubits.

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1. Introduction

The term of "quantum spin dynamics" is now commonly used to qualify quantum relaxation phenomena issued from slow and incoherent spin reversal by resonant tunneling with very small tunneling splitting Δ. The first example, given by the "macroscopic quantum tunneling" of an ensemble of $S = 10$ molecule spins (Mn_{12}-AC), forming a single crystal of nanomagnets [1], has been followed by similar studies in another $S = 10$ molecule (Fe_8), and then by the discovery of a large number of molecules with similar behavior (single molecule magnets, SMM). The most important characteristic of these molecules is a very small tunneling gap resulting from their large spin. In fact, in order to connect the states S and $-S$ by e.g. a transverse field $H_\perp$, the operator S_+ has to be applied $2S$ times (S must be integer), showing that $\Delta \sim (H_\perp/H_A)^{2S}$, where $H_A = 2DS/g\mu_B \geqslant H_\perp$ is the anisotropy field. This property of spin operators leads to similar expressions in zero-field when $H_\perp$ (1st order) is replaced by higher-order transverse anisotropy parameters (2nd, 4th ...). These magnetic systems with very small tunneling gaps show the coexistence of incoherent quantum relaxation and classical hysteresis loop, as in their archetype Mn_{12}-AC. The reason for that is very simple: (i) the tunneling gap being smaller than inhomogeneous level broadening by environmental fluctuations (phonons, other spins, free

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